Hazardous Air Pollution from Mobile Sources: A Comparison of Alternative Fuel and Reformulated Gasoline Vehicles

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ABSTRACT

Although there have been several studies examining emissions of criteria pollutants from in-use alternative fuel vehicles (AFVs), little is known about emissions of hazardous air pollutants (HAPs) from these vehicles. This paper explores HAP tailpipe emissions from a variety of AFVs operating in the federal government fleet and compares these emissions to emissions from identical vehicles operating on reformulated gasoline. Emissions estimates are presented for a variety of fuel/model combinations and on four HAPs (acetaldehyde, 1,3-butadiene, benzene, and formaldehyde). The results indicate that all AFVs tested offer reduced emissions of HAPs, with the following exceptions: ethanol fueled vehicles emit more acetaldehyde than RFG vehicles, and ethanol- and methanol-fueled vehicles emit more formaldehyde than RFG vehicles. The results from this paper can lead to more accurate emissions factors for HAPs, thus improving HAP inventory and associated risk estimates for both AFVs and conventional vehicles.

IMPLICATIONS

Alternative fuel vehicles are becoming more prevalent in urban areas and are likely to see increased market penetration due to incentives and mandates in the Clean Air Act and the Energy Policy Act. Because little is known about emissions of HAPs from these vehicles and about how these emissions compare to conventional gasoline vehicles, policymakers find it difficult to determine the overall health impacts of AFV introduction. This paper quantifies emissions of four important HAPs for a variety AFVs and compares these emissions with those from similar vehicles operating on reformulated gasoline. Emissions factors generated from these results can be used to improve HAP inventories and associated risk estimates for AFVs and conventional vehicles.

INTRODUCTION

Emissions from mobile sources represent an ever-increasing share of the total criteria pollution inventory in urban areas. In 1995, highway vehicles contributed 63.6% of total carbon monoxide, 34.9% of total nitrogen oxides, and 26.7% of total volatile organic compound emissions nationally.¹ Although many steps have been taken in recent years to control these pollutants, increases in vehicle miles traveled, vehicle populations, consumer tampering, poor vehicle maintenance, and changing driving patterns have hampered these efforts.^{2,3}

In the past, command-and-control, technologybased standards have been the mechanism of choice for mobile source regulators. But with the passage of the Clean Air Act Amendments of 1990 (CAAA) and the Energy Policy Act of 1992 (EPACT), the option of using clean, alternative fuels has emerged as a possible solution to mobile source air pollution. Oxygenated and reformulated fuels have shown promise in reducing criteria pollutants, and programs requiring the use of these fuels were explicitly outlined in the CAAA.^{4,5} However, oxyfuels and reformulated gasoline (RFG) are not considered "alternative fuels" under the definition outlined in EPACT, a piece of legislation that also sought to reduce U.S. reliance on imported petroleum. To meet the goals of EPACT, vehicles must operate on "substantially non-petroleum based fuels" such as natural gas, methanol, ethanol, propane, and electricity. EPACT identifies a variety of mandates that require fleet operators in metropolitan areas to begin purchasing vehicles that run on these alternative fuels.⁶

Although alternative fuel vehicles (AFVs) currently make up only a small percentage of the U.S. vehicle population, it is expected that these vehicles will begin to penetrate markets at significant levels in the near future. The number of AFVs that were in operation in the United States in 1996 was about 352,000, but this number is expected to increase to over 400,000 by 1998 and to over 4 million by 2005.^{7,8} Also, due to AFV credit markets that are emerging in response to EPACT, a majority of these 4 million vehicles may be centered in a few large urban areas.⁹

There have been an increasing number of studies over the past several years that attempt to understand better the emissions characteristics of these AFVs.^{10–13} Most of these studies suggest that AFVs are cleaner than conventional vehicles operating on reformulated gasoline. As of yet, however, very little has

been done in the area of quantifying hazardous air pollutant (HAP) emissions from these AFV sources. The objective of this paper is to quantify and compare HAP emissions between AFVs and conventional vehicles. This paper focuses on emissions of 1,3-butadiene, acetaldehyde, benzene, and formaldehyde. The U.S. Enviromental Protection Agency (EPA) (through Section 112 of the CAAA) has identified these pollutants in a list of 40 HAPs that present significant risks to human health in urban areas.

Of the 40 HAPs identified by EPA, the 4 analyzed here are largely emitted by mobile sources. EPA estimates that 60% of total benzene emissions, 94% of 1,3-butadiene emissions, 39% of acetaldehyde emissions, and 33% of formaldehyde emissions are from mobile sources.¹⁴ Table 1 shows EPA's estimates of the total annual mobile source emissions and average emissions factor (mg/mi) for each of these pollutants. The emissions factors are determined for several years and are based on assumptions of vehicle use and fuel type. EPA is using these estimates as supporting data for their proposed HAP regulatory strategy. Yet EPA has recognized that because more AFVs will be added to urban fleets over the next few years, "the potential cancer reduction benefits resulting from the combustion of these alternative fuels should be addressed."14

Existing HAP emission estimates from AFVs are limited due to the small-scale studies under which they were estimated.¹⁵⁻¹⁹ Because of the limited number of vehicles under study, previous estimates of HAP emissions factors for AFVs are not statistically reliable. This paper addresses this dearth of data analysis by exploring emissions from an extensive data set collected by the U.S. Alternative Fuels Data Center (AFDC).

ANALYSIS AND RESULTS

Part of the reason more rigorous statistical studies have not been performed on AFV emissions is that the data are sparse. One organization has attempted to address

Table 1. National mobile source HAP emissions inventory for 1990.

НАР	Emissions (tons/yr)	Emissions Factor, 1990 Base (mg/mi)	Emissions Factor, 1995 Base (mg/mi)	Emissions Factor, 2000 Base (mg/mi)	
Benzene	208,740	88.2	47.2	35.1	
1,3 Butadiene	36,920	15.6	9.4	7.1	
Fomaldehyde	97,506	41.2	23.4	16.2	
Acetaldehyde	28,163	11.9	7.1	5.1	

Source: EPA, 1993.

this lack of high quality AFV emissions data, however. The National Renewable Energy Lab (NREL), under funding from the U.S. Department of Energy, has been collecting emissions data since the early 1990s on more than 300 AFVs and RFG control vehicles operating in the federal vehicle fleet. These vehicles operate on a variety of fuels, including methanol blends, ethanol blends, compressed natural gas, and propane. (Note that RFG represents California Phase II Certification fuel). Vehicles are operated in various federal agency fleets and represent a variety of driving conditions and operations.

Emissions tests on these vehicles are conducted at three labs certified by EPA. Each vehicle is tested using the EPA's Urban Dynamometer Driving Schedule of the Federal Test Procedure protocol at odometer readings of approximately 4,000 mi, 10,000 mi, and every 10,000 mi thereafter. Data are reported on the weighted FTP test results. The general test procedures, emissions test driving profiles, and other facts about this program are reported in other publications.¹¹⁻¹³

For each vehicle test, NREL collects emissions data on standard criteria pollutants and ozone precursors (namely, carbon monoxide, nitrogen oxides, total hydrocarbons, and non-methane hydrocarbons). In addition, a vast majority of the vehicles undergo formaldehyde and acetaldehyde testing via impinger extraction and analysis using a liquid chromatograph. A smaller number of vehicles undergo testing for benzene and 1,3-butadiene emissions (as well as other speciated hydrocarbons) via gas chromatography.

This paper uses data acquired on January 17, 1998, from the NREL dataset. Data were obtained for vehicles identified in Tables 2–5. These vehicles represent a variety of makes (Ford, Dodge, General Motors) and models (e.g., Spirit, Intrepid, Lumina, Ram Van, Taurus). All the M85 and E85 vehicles are flexible-fuel vehicles and are tested on both alternative and conventional fuels. Fuels analyzed in this study include compressed natural gas (CNG), 85% methanol blend (M85), and 85% ethanol blend (E85).

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Table 2. Emissions results for 1,3-butadiene.

Model	Test Fuel	Model Year	N	Mean Emissions (mg/mi)	Mean NMHC Emissions (g/mi)	Weight Percent of NMHC (%)	Mean Odometer (mi)	<i>t</i> test for AFV-RFG Comparison
GM Intrepid	M85	1995	6 (9)	0.11 (0.02)	0.114	0.95%	9237	-9.76 (<0.0001)*
GM Intrepid	RFG	1995	10 (17)	0.79 (0.17)	0.113	0.66%	9749	
Dodge Ram Van	CNG	1992	3 (5)	0.12 (0.13)	0.271	0.05%	14176	-15.76 (<0.0001)*
Dodge Ram Van	RFG	1992	4 (8)	2.03 (0.17)	0.276	0.73%	13694	
Dodge Spirit	M85	1993	11 (16)	0.23 (0.29)	0.113	0.20%	16387	-2.70 (0.012)*
Dodge Spirit	RFG	1993	20 (32)	0.68 (0.50)	0.097	0.70%	18235	
Ford Taurus	E85	1995	6 (9)	0.18 (0.04)	0.096	0.19%	10406	-12.68 (<0.0001)*
Ford Taurus	RFG	1995	11 (18)	0.56 (0.07)	0.090	0.62%	10279	

Note: *N* represents the number of vehicles in the sample. For *N*, the number in parentheses represents the total number of data points (i.e., tests) for the vehicle sample. For mean HAP emissions, the number in parentheses represents the standard deviation.

Table 3. Emissions results for acetaldehyde.

Model	Test Fuel	Model Year	N	Mean Emissions (mg/mi)	Mean NMHC Emissions (g/mi)	Weight Percent of NMHC (%)	Mean Odometer (mi)	<i>t</i> test for AFV-RFG Comparison
Econoline Van	M85	1992	13 (24)	0.40 (0.20)	0.069	0.58%	18809	-3.92 (0.0006)*
Econoline Van	RFG	1992	13 (24)	0.86 (0.37)	0.167	0.51%	18816	
Chevy Lumina	E85	1992	13 (25)	26.40 (5.44)	0.086	7.56%	13859	16.84 (<0.0001)*
Chevy Lumina	RFG	1992	13 (28)	1.00 (0.01)	0.165	0.61%	13764	
Chevy Lumina	E85	1993	12 (33)	15.52 (3.45)	0.082	18.93%	23698	22.69 (<0.0001)*
Chevy Lumina	RFG	1993	28 (76)	0.89 (0.28)	0.173	0.51%	17960	
GM Intrepid	M85	1995	25 (45)	0.20 (0.03)	0.089	0.22%	8943	-14.71 (<0.0001)*
GM Intrepid	RFG	1995	49 (92)	0.48 (0.09)	0.129	0.37%	9607	
Dodge Ram Van	CNG	1992	37 (109)	0.32 (0.22)	0.126	0.25%	12132	-9.42 (<0.0001)*
Dodge Ram Van	RFG	1992	22 (60)	1.30 (0.56)	0.300	0.43%	19406	
Dodge Ram Van	CNG	1994	16 (36)	0.32 (0.23)	0.052	0.62%	11222	-6.58 (<0.0001)*
Dodge Ram Van	RFG	1994	33 (85)	1.53 (0.72)	0.336	0.46%	25051	
Dodge Spirit	M85	1993	77 (139)	0.32 (0.24)	0.058	0.55%	14230	-3.56 (0.0005)*
Dodge Spirit	RFG	1993	150 (286)	0.51 (0.47)	0.130	0.39%	15890	
Ford Taurus	E85	1995	22 (38)	11.84 (5.58)	0.109	10.86%	9902	14.00 (<0.0001)*
Ford Taurus	RFG	1995	45 (82)	0.29 (0.12)	0.103	0.28%	8987	

Note: *N* represents the number of vehicles in the sample. For *N*, the number in parentheses represents the total number of data points (i.e., tests) for the vehicle sample. For mean HAP emissions, the number in parentheses represents the standard deviation.

Model	Test Fuel	Model Year	N	Mean Emissions (mg/mi)	Mean NMHC Emissions (g/mi)	Weight Percent of NMHC (%)	Mean Odometer (mi)	<i>t</i> test for AFV-RFG Comparison
GM Intrepid	M85	1995	6 (9)	1.05 (0.14)	0.086	1.22%	9237	-10.86 (<0.0001)*
GM Intrepid	RFG	1995	10 (17)	4.16 (0.68)	0.130	3.21%	9749	
Dodge Ram Van	CNG	1992	3 (5)	0.46 (0.46)	0.241	0.19%	18153	-33.35 (<0.0001)*
Dodge Ram Van	RFG	1992	4 (8)	11.63 (0.49)	0.277	4.20%	13694	
Dodge Spirit	M85	1993	11 (16)	1.91 (1.05)	0.063	3.03%	16387	-2.32 (0.028)*
Dodge Spirit	RFG	1993	20 (32)	4.60 (3.74)	0.131	3.50%	18235	
Ford Taurus	E85	1995	6 (9)	1.18 (0.17)	0.096	1.23%	10406	-12.42 (<0.0001)*
Ford Taurus	RFG	1995	11 (18)	3.02 (0.34)	0.088	3.45%	10279	

 Table 4. Emissions results for benzene.

Note: N represents the number of vehicles in the sample. For N, the number in parentheses represents the total number of data points (i.e., tests) for the vehicle sample. For mean HAP emissions, the number in parentheses represents the standard deviation.

For 1,3-butadiene and benzene, model/fuel combinations with greater than three vehicles in their population were selected. For acetaldehyde and formaldehyde, model/fuel combinations with greater than 10 vehicles in their population were selected. (This difference is due to the fact that fewer vehicles were tested for 1,3-butadiene and benzene, and so vehicle samples were smaller). On average, there were approximately 15 vehicles per sample, with the smallest sample having three vehicles and the largest having 150 vehicles. It is recognized that the small populations of some of the vehicle samples will affect the significance of the analysis; however, even low population combinations will provide some insight into HAP emissions from AFVs.

The methodology used for this paper is straightforward. For each vehicle and HAP, an average emissions factor was calculated from all of the tests that were conducted on that vehicle. With some exceptions, most vehicles have undergone one or two emissions tests. These data were then pooled into samples that had identical make, model, and model year vehicles. For a given make, model, and model year, *t* tests were conducted to compare emissions from vehicles operating on different fuels. From the *t* tests, differences in HAP emissions from different fuels could be identified at a statistically significant level (i.e., at a 95% confidence level). Thus, the null hypothesis tested is that there is no difference in HAP emissions from AFVs and conventional vehicles.

The results of the analyses are presented in Tables 2–5 for each of the HAPs under study. For each table, an average emissions factor is presented (in mg/mi)

for the HAP, along with an average odometer reading for the sample. In addition, we include the average non-methane hydrocarbon (NMHC) emissions for each sample, as well as the HAP percent of NMHC emissions by mass. Identical vehicles that operate on different fuels are grouped in consecutive rows. The pvalues from the t tests are also shown. Tests that permit us to reject the null hypothesis with 95% confidence are noted with an asterisk.

Although more data will help in conducting similar analyses in the future, the results of this set of analyses allow some important conclusions to be drawn. These are highlighted below:

- Every AFV shows significantly lower emissions for 1,3-butadiene when compared to RFG vehicles. In addition, RFG values presented here are much lower than EPA estimates (see Table 1). This may be due to the fact that these RFG vehicles were tested on California Phase II RFG, while EPA estimates are based on estimates of the national vehicle population and fuel characteristics.
- Every AFV shows significantly lower emissions for acetaldehyde, except E85 vehicles, which show considerably higher acetaldehyde emissions. Again, EPA estimates (see Table 1) are high in comparison to the RFG results, for the possible reasons stated above.
- Similar to 1,3-butadiene, every AFV shows significantly lower emissions for benzene when compared to RFG vehicles. As above, the EPA estimates for benzene (see Table 1) are high compared to these results.

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Table 5. Emissions results for formaldehyde.

Model	Test Fuel	Model Year	N	Mean Emissions (mg/mi)	Mean NMHC Emissions (g/mi)	Weight Percent of NMHC (%)	Mean Odometer (mi)	<i>t</i> test for AFV-RFG Comparison
Econoline Van	M85	1992	13 (24)	19.66 (9.37)	0.069	28.63%	18809	5.21 (<0.0001)*
Econoline Van	RFG	1992	13 (24)	5.10 (3.71)	0.167	3.05%	18816	
Chevy Lumina	E85	1992	13 (25)	7.80 (2.45)	0.086	9.07%	13859	4.29 (0.0003)*
Chevy Lumina	RFG	1992	13 (28)	4.51 (1.29)	0.165	2.73%	13764	
Chevy Lumina	E85	1993	12 (33)	4.78 (1.22)	0.082	5.86%	23698	0.53 (0.599)
Chevy Lumina	RFG	1993	28 (76)	4.54 (1.41)	0.173	2.61%	17960	
GM Intrepid	M85	1995	25 (45)	16.86 (1.59)	0.089	18.99%	8943	61.33 (<0.0001)*
GM Intrepid	RFG	1995	49 (92)	2.04 (0.43)	0.129	1.58%	9607	
Dodge Ram Van	CNG	1992	37 (109)	5.16 (3.95)	0.126	4.12%	12132	1.19 (0.237)
Dodge Ram Van	RFG	1992	22 (60)	4.13 (1.05)	0.300	1.38%	19406	
Dodge Ram Van	CNG	1994	16 (36)	3.94 (3.41)	0.052	7.61%	11222	-2.95 (0.005)*
Dodge Ram Van	RFG	1994	33 (85)	7.30 (3.88)	0.336	2.17%	25051	
Dodge Spirit	M85	1993	77 (139)	12.13 (3.15)	0.058	20.74%	14230	34.93 (<0.0001)*
Dodge Spirit	RFG	1993	150 (286)	1.93 (1.19)	0.130	1.48%	15890	
Ford Taurus	E85	1995	22 (38)	2.60 (0.99)	0.109	2.39%	9902	8.96 (<0.0001)*
Ford Taurus	RFG	1995	45 (82)	1.13 (0.35)	0.103	1.09%	8987	

Note: *N* represents the number of vehicles in the sample. For *N*, the number in parentheses represents the total number of data points (i.e., tests) for the vehicle sample. For mean HAP emissions, the number in parentheses represents the standard deviation.

• Only one CNG sample shows lower emissions estimates for formaldehyde. Both alcohol fuels (E85 and M85) show higher formaldehyde emissions when compared to RFG vehicles. Again, EPA formaldehyde emissions estimates (see Table 1) are high when compared to the results presented here.

CONCLUSIONS

The data analyses presented in this paper are a first attempt to begin to understand HAP emissions from AFVs. Since AFVs are likely to become more prevalent in the U.S. transportation sector over the next decade, environmental policymakers and researchers need to understand the impacts of AFVs on toxic air emissions. These emission estimates must be incorporated into larger comparative risk assessments that analyze the increases or decreases of cancer due to toxic emissions from AFVs.

Of course, a number of caveats exist when analyzing a relatively small set of vehicle emissions data. One must be concerned about whether the data truly represent vehicle populations as a whole. One also must address emissions deterioration over the life of the vehicle, an issue not considered explicitly in this paper. Thus, interpreters of the analytical results presented here should bear in mind the sample sizes and odometer readings of each individual test before drawing strong conclusions.

The next steps to extend this preliminary research include applying an emissions deterioration model to determine how HAP emissions deteriorate over the lifetime of a vehicle. Using such a model, one can determine the lifetime vehicle emissions of HAPs for various fuels. These lifetime estimates can then be used in modeling exercises to explore the potential impacts that AFVs might have on HAP inventories. Finally, using these newly generated HAP inventories, one can apply exposure assessments to determine the decreased (or increased) health risks that AFVs may have on urban populations.

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REFERENCES

- National Air Pollutant Emission Trends; U.S. Environmental Protection Agency. U.S. Government Printing Office: Washington, DC, 1996.
- Black, F.M. "Control of motor vehicle emissions—The U.S. experience," Crit. Rev. in Environ. Control. 1991, 21, 373-410.
- LeBlanc, D.C.; Saunders, F.M.; Meyer, M.D.; Guensler, R. "Driving pattern variability and impacts on vehicle carbon monoxide emissions," *Transportation Res. Record* 1995, *1472*, 45-52.
 Stump, F.D.; Knapp, K.T.; Ray, W.D.; Siudak, P.D.; Snow, R.F. "Influence of component for the set of set of
- Stump, F.D.; Knapp, K.T.; Ray, W.D.; Siudak, P.D.; Snow, R.F. "Influence of oxygenated fuels on the emissions from three pre-1985 light-duty passenger vehicles," J. Air & Waste Manage. Assoc. 1994, 44, 781-786.
- Kirchstetter, T.W.; Singer, B.C.; Harley, R.A.; Kendall, G.R.; Chan, W. "Impact of oxygenated gasoline use on California light-duty vehicle emissions," *Environ. Sci. Technol.* 1996, *30*, 661-670.
- Winebrake, J.J. "Another challenge for energy/environmental strategists: Federal laws affecting vehicle fleets," *Strategic Planning for Energy and the Environ.* 1994, 13, 52-67.
- Alternatives to Traditional Transportation Fuels 1996; U.S. Department of Energy. U.S. Government Printing Office: Washington, DC, 1997; DOE/EIA-0585(94)/1.
- Annual Energy Outlook, 1994; U.S. Department of Energy. U.S. Government Printing Office: Washington, DC, 1994.
- Winebrake, J.J.; Farrell, A.E. "The AFV credit program and its role in future market development," *Transportation Res.: Part D—Transport* and the Environ. 1997, 2, 125-132.
- Gabele, P. "Exhaust emissions from in-use alternative fuel vehicles," J. Air & Waste Manage. Assoc. 1995, 45, 770-777.
- Kelly, K.J.; Bailey, B.K.; Coburn, T.C.; Clark, W.; Eudy, L.; Lissiuk, P. "FTP emissions test results from flexible-fuel methanol Dodge Spirits and Ford Econoline vans," SAE Technical Paper Series 1996, 961090, 207-230.
- Kelly, K.J.; Bailey, B.K.; Coburn, T.C.; Eudy, L.; Lissiuk, P. "Round 1 emissions test results from compressed natural gas vans and gasoline controls operating in the U.S. federal fleet," *SAE Technical Paper Series* 1996, 961091, 233-248.
- Kelly, K.J.; Bailey, B.K.; Coburn, T.C.; Clark, W.; Lissiuk, P. "Federal test procedure emissions test results from ethanol variable-fuel vehicle Chevrolet Luminas," SAE Technical Paper Series 1996, 961092, 249-268.
- 14. *Motor Vehicle-Related Air Toxics Study*; U.S. Environmental Protection Agency, Office of Mobile Sources. U.S. Government Printing Office: Washington, DC, 1993; EPA-420-R-93-005.

- Gabele, P. "Characterization of emissions from a variable gasoline/methanol fueled car," J. Air & Waste Manage. Assoc. 1990, 40, 296-304.
- Definition of a Low-Emission Motor Vehicle in Compliance with the Mandates of Health and Safety Code Section 39037.05; California Air Resources Board. Mobile Source Division. CARB: El Monte, CA, 1989.
- Proposed Reactivity Adjustment Factors for Transitional Low-Emission Vehicles: Technical Support Document; California Air Resources Board. Mobile Source Division. CARB: El Monte, CA, 1991.
- Emissions and Air Quality Modeling Results from Methanol/Gasoline Blends in Prototype Flexible/Variable Fuel Vehicles; Technical Bulletin No. 7; Auto/Oil Air Quality Improvement Research Program. Coordinating Research Council: Atlanta, GA, 1992.
- Emissions of Three Dedicated-Methanol Vehicles; Technical Bulletin No. 10; Auto/Oil Air Quality Improvement Research Program. Coordinating Research Council: Atlanta, GA, 1992.

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